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LIMITATIONS IN THE USE OF THE END-WINDOW COUNTER IN THE ASSAYING OF STREAM WATERS FOR BETA RADIOACTIVITY

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LIMITATIONS IN THE USE OF THE END-WINDOW COUNTER IN THE ASSAYING OF STREAM WATERS FOR BETA RADIOACTIVITY

Stream water to be assayed for beta radioactivity will have natural radioactivity with or without artificial radioactivity. The principle source of natural radioactivity is from K^{40} and from uranium, thorium and their daughter products. The artificial radioactivity will be the result of contamination from (a) the disposal of radioactive wastes, (b) fall-out from a contaminated atmosphere, and (c) the use of stream waters by atomic energy installations. The assay is made primarily to determine the gross amount of radioactivity present. When the level of radioactivity in water is in the order of 10 micromicrocuries per liter, it is increasingly difficult to identify the individual radioactive isotopes by the usual methods of half-life and energy determinations.

The radioactivity of a sample is expressed in terms of disintegrations per unit of time. Since an end-window counter does not detect all of the disintegrations in a sample, the observed radioactivity in counts per unit of time is divided by the efficiency of the counter to give the "absolute" or true beta radioactivity. The overall efficiency of a counter depends on its geometry and factors of backscatter, air and window absorption, self-absorption, technique of mounting the sample, etc.

The literature [1,2,3,15,17] discusses various methods of determining the absolute beta radioactivity with end-window counters under specified conditions. However, many of these specified conditions cannot

be met in the absolute beta assay of stream water. Therefore, it has been necessary to determine each of the factors contributing to the overall efficiency. This paper will discuss the use of the end-window counter for the assay of the absolute beta radioactivity of stream water, its limitations, and the necessary corrections.

Following the end-window counter studies, internal proportional (gas flow) counters were studied. The main advantages of some internal counters are that (a) larger samples can be evaporated on dishes without increasing self-absorption, (b) the counting efficiency is improved by the elimination of air and window absorption, and (c) the geometry of counting is increased to 2 pi or 50 percent. As a consequence, internal proportional counters have been tested and adopted by the Environmental Health Center for the "absolute" counting of low level radioactivity in water. Reports on the application of internal counters for this purpose are in preparation.

Counter and Mount

The counting arrangement of the particular end-window tube and sample mount used for our investigation is shown in Figure 1. The counter had a window thickness of 1.8 mg/cm^2 and a window diameter of 2.8 cm.

Sample

The low levels of beta radioactivity which occur in stream water make it necessary to evaporate relatively large volumes of sample for a good statistical evaluation[8] within a practical counting time of about 30 minutes. Frequently the dry solids of the sample are so high that

they introduce significant losses of radiation by self-absorption(11). These losses can be reduced by spreading the dry solids in a thin film over a large area providing all of the radioactivity is counted with the same efficiency. Usually end-window G-M tubes have a diameter less than 3 cm. and the sample dish or planchet should have a smaller diameter for efficient counting of an extended source.

Geometry

The geometry of end-window counters is defined as the ratio of the solid angle subtended by the sensitive counter volume with respect to the source to the total (4 pi) solid angle. The value of the ratio of the angles can be calculated for point source(17) and has been determined experimentally(1,2,17) for both point and extended sources. This geometry is not identical with the overall counting efficiency of the counter.

The geometry of a point source is approximately 16 percent for the first shelf of the mount shown in Figure 1. However, dishes having a height of 1 cm. do not fit on the first shelf of our instrument. Sample dishes of height greater than 1 cm. can be placed on the second shelf, but here the geometry of a point source is only 5.4 percent. Gleason et al(6) placed a metal plate below the window of the G-M tube with an orifice of diameter less than the window diameter. With this arrangement the insensitive regions of the counting tube are avoided and a more accurate and reproducible geometry can be determined. When this technique was applied to the counting arrangement in Figure 1, the geometry of a point source on the second shelf was found to be only 3.25 percent.

When the sample solids are spread over the area of a planchet or dish, the geometry of a point source no longer applies. According to Cook et al[2] the geometry of such an extended source is less than the point source geometry. They minimized the reduction of the geometry by placing the sample very close to the window (where the geometry of a point source is greater than 40 percent), or an appreciable distance from the window (where geometry of a point source is less than 5 percent), and by using a counter with a window diameter larger than the diameter of the sample.

An experiment was performed to arrive at the geometry of a source when extended to nearly twice the diameter of the counter window. A source of P^{32} one millimeter in diameter was successively counted on a shelf as it was moved from a point directly beneath the center of the window to one side in steps of 2 mm. The test was made on each of three shelves. The results are presented in Figure 2 and show that as the point source is moved to the periphery of a dish or planchet, there is an increasing loss of counting efficiency. For example, if the diameter of the dish is 1.5 times that of the window, a point source on the second shelf will count less than one-half as much at the periphery as it would in the center.

The results in Figure 2 were used to summate the efficiency of counting a radioactive source spread uniformly over the entire area of a dish, as compared to the count of an equivalent point source. The results are presented in Figure 3 and are in agreement with predictions made by Burtt[1]. Figure 3 shows, for example, that a source placed on the first shelf

directly under the window and uniformly spread over a dish diameter equal to the window would count about 80 percent of the activity of an equivalent point source in the center. If the sample solids are uniformly spread over the area of an aluminum dish 4.8 cm. in diameter with a capacity of 14 ml. (such as we have used for internal counters), the end-window counter would record only 65 percent of an equivalent point source at the center of the dish. Thus, the net geometry of this extended source is 5.4 x 0.65 or only 3.5 percent.

Backscatter[1,12,16,17]

When radiations emitted in a direction opposite to the window are deflected by the sample dish and shelf into the window, it is called backscatter. When a source is mounted on a backing of negligible thickness, such as a very thin film of Formvar plastic[5] and counted, the backscatter is negligible. When the same source on the film is now placed on a planchet or dish, a higher counting rate will be observed. The difference in counting rate is due to backscattered radiation. The ratio of the latter to the former counting rate is the backscatter factor which is a part of the overall counter efficiency. Backscatter increases with an increase in the atomic number and thickness of the sample backing and reaches a maximum or saturation value at a backing thickness which corresponds to about one-fifth of the range of the radiation[4].

In the process of being backscattered, the radiation loses energy and the softer radiation is more strongly absorbed by the air and window. Consequently, there is a decrease in the apparant saturation backscatter.

particularly for low energy emitters[17]. It has been shown[17] that if correction for this absorption is taken into account, the saturation backscatter is independent of energy over the range 0.3 to 1.7 Mev.

The backscatter factor will vary slightly with the geometry[12,17] because the backscatter is not isotropic. For P^{32} mounted on an aluminum dish and placed on a thick copper support, the backscatter factor corrected for air + window absorption was found to be 1.41.

Air + Window Absorption[17]

Some radiation is absorbed in passing from a sample source through a column of air and the G-M window. The amount absorbed is dependent on the energy of the radiation[9] and the mass of the absorbing medium whose thickness is usually expressed in milligrams per square centimeter. By counting the radiation passing through different thicknesses of standard aluminum absorbers, it is possible to extrapolate the absorption curve from zero aluminum absorber to a thickness equivalent to the air and window. The air + window thickness of a sample placed on the second shelf in Figure 1 is equivalent to about 5 mg/cm^2.

The procedure of extrapolating to zero absorber thickness is subject to considerable uncertainty, particularly for low energy beta emitters. An appreciable fraction of the radiation from P^{32}, I^{131} and Ca^{45} would be completely absorbed by a thickness equivalent to the air and window, according to internal counter studies[7,13]. Thus Zumwalt[17] found that when the absorption curve of the relatively soft emitter I^{131}

Air Scatter (17)

Air scatter produces an increase in counting efficiency but becomes negligible when the source is very close to the counter window. For the first two shelves of Figure 1, air scatter may be estimated to be about 2 percent for soft emitters, such as Co^{60} (max. energy 0.31 Mev.) and negligible for beta particles of higher energy.

Counting Efficiency

The absolute radioactivity, D, may be determined from the observed radioactivity, I, by means of the appropriate corrections according to the equation,

$$D = \frac{I}{GBT} \qquad (1)$$

where G = geometry factor

B = backscatter factor,

and T = transmission factor which is the product of the transmission factors due to air + window absorption and self-absorption.

The counting efficiency, E, is the ratio of the observed radioactivity to the absolute radioactivity or from Eq. (1).

$$E = \frac{I}{D} = GBT \qquad (2)$$

Since $\Theta \leq 1$, $1 \leq B \leq 2$, $T \leq 1$, it is apparent from Eq. (2) that the counting efficiency is highest when B is as large as possible, $\Theta = 1$, and $T = 1$. The counting efficiency of the end-window counter in Figure 1 for an extended source counted in an aluminum dish on the second shelf is

$$E = 0.035 \times 1.41\, T = 0.049T \qquad (3)$$

The maximum counting efficiency with negligible absorption is 4.9 percent. With a soft emitter the window absorption may reduce the counting efficiency to zero.

If one assumed that unknown energies are greater than 0.15 Mev., then the transmission factor may be estimated. The average energy for 0.15 Mev. (max.) will be about 0.05 Mev., and the air + window thickness of 5 mg/cm^2 will absorb at least 50 percent of the radiation. Actually more than 50 percent of the radiation will be absorbed because the energy frequency distribution of the beta spectrum is skewed positive. Assuming a rough estimate of 60 percent absorption, the transmission factor would then be 0.40.

For the higher energies, on the other hand, the transmission factor may be estimated to be 1.00. Assuming a mean estimate of $T = 0.70$, the error in T will not be greater than 43 percent providing self-absorption is negligible.

Low Level Counter Statistics

When low levels of radioactivity (less than the background of the instrument) are measured, the error due to counting statistics becomes large for a counting time of 32 minutes. Consider a normal background of 80 cpm for an end-window counter in a lead shield and the maximum permissible water contamination[10] of 100 μμc/liter or 222 dpm/liter. The usual sample taken to minimize self-absorption is about 200 ml. At a level of 100 μμc/liter this sample will have 44 dpm. With a counting efficiency of 4.9 percent, the observed net count is only 2.2 cpm. For a counting time of 32 minutes the net count will have a statistical counting error of ± 2.2 cpm or ± 100 percent of the net count at a confidence level of 95 percent.

If the transmission factor is 0.7, the net counting efficiency is 3.4 percent, or a 200 ml. sample containing 100 μμc/liter will be counted as 1.5 ± 2.2 counts per minute.

The geometry of the counter for samples mounted in dishes 48 mm. in diameter and 8 mm. in height may be improved by placing the dish as close to the window as possible. The resulting geometrical efficiency for an extended source 10 mm. from the window will be 6.5 to 9.2 percent, depending on the window to sensitive volume distance. Assuming a geometrical efficiency of 6.5 percent, the net counting efficiency becomes 6.4 percent. The observed radioactivity of a 200 ml. sample at the 100 μμc/l. level will then be 2.8 ± 2.3 cpm at an assumed T value of 0.7.

SUMMARY

The counting efficiency of an end-window counter is in the order of 3.5 percent when a sample, evaporated in an aluminum dish 48 mm. in diameter and 0.8 cm. in height, is counted on the second shelf of the counter used in our studies. These dishes have a capacity of 14 ml. and are particularly suitable for evaporating large water samples directly within the dish and counting in large internal proportional counting chambers. The use of smaller diameter dishes or planchets for mounting samples may result in greater self-absorption losses but they are to be preferred for end-window counting since a counting efficiency of as much as 25 percent (subject to considerable uncertainty) may be achieved.

The backscatter factor for the aluminum dish on a copper support, corrected for air + window absorption is in the order of 1.4.

Air + window absorption and self-absorption are energy dependent.

Self-absorption is negligible if the sample thickness is not more than 0.5 mg/cm^2.

Air scatter may be estimated to be about 2 percent on the first two shelves for soft emitters and negligible for the higher energies.

With isotopes of known energies, the air + window transmission is uncertain by the extrapolation method. The error may be as much as 30 percent. With unknown energies the transmission value cannot be determined, but may be estimated to be 0.70 ± 0.30.

The overall counting efficiency of an end-window counter for a sample mounted in a dish 48 mm. in diameter and 0.8 cm. in height is usually less than 6.4 percent, primarily because the geometry is low. When used for the assay of unknown energies at low levels, the assay of absolute radioactivity is unreliable, due to the large error in estimating the transmission through the air + window thickness.

At the maximum permissible water contamination of 100 μμc/liter the statistics of counting for a practical counting time of 30-32 minutes introduce a large error which, at the 95 percent confidence limit, may amount to more than 80 percent of the cpm/200 ml. of sample.

CONCLUSIONS

The use of the end-window counter is limited for assaying the beta radioactivity of stream water at levels in the order of 100 μμc/l. (10^{-7} μc/ml.). Its poor geometry makes it relatively insensitive to very low levels of radioactivity. The air + window absorption renders it insensitive to low energy radiation. The counter can only evaluate absolute radioactivity with a very large error. This is true if the energies of the radioactive contaminants are known or unknown. The end-window counter can at best indicate the presence of radioactivity at the maximum contamination level of water.

FIGURE 1

G-M TUBE AND MOUNT ASSEMBLY

Distances from Window to Bottom of Shelves

Shelf No.	Distance cm.
1	0.87
2	2.37
3	3.92
4	5.52
5	7.17
6	8.72

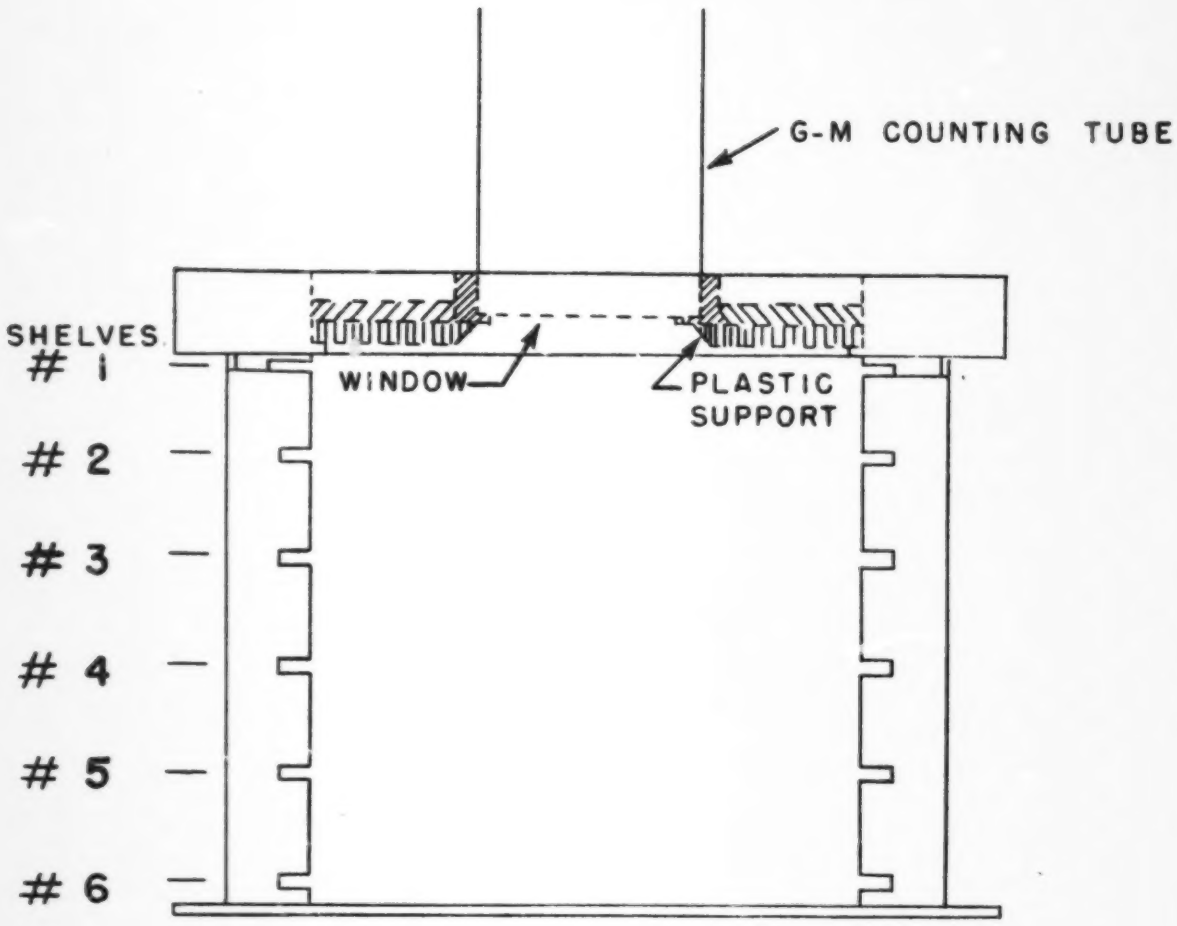

G-M COUNTING TUBE
SHELVES
1
2
3
4
5
6
WINDOW
PLASTIC SUPPORT

FIGURE 2

The relative counting rate of a point source displaced at right angles to the axis of the G-M tube.

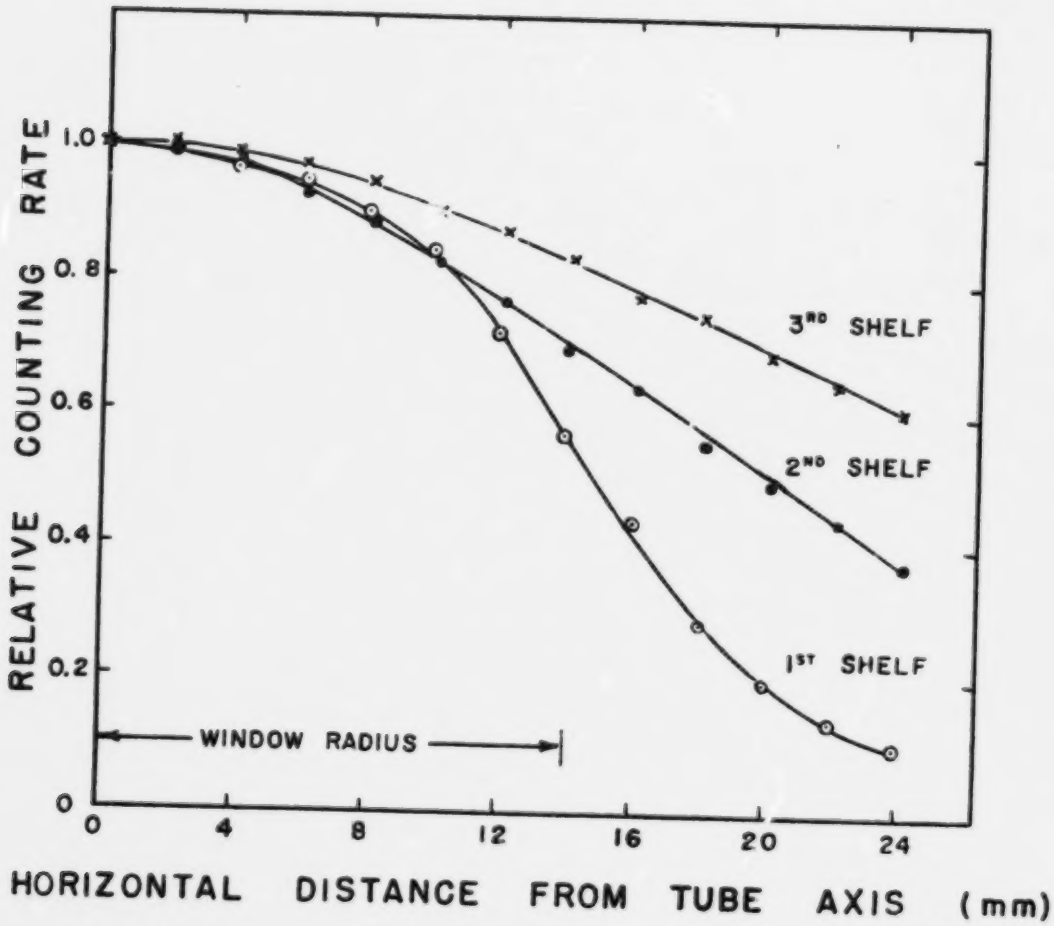
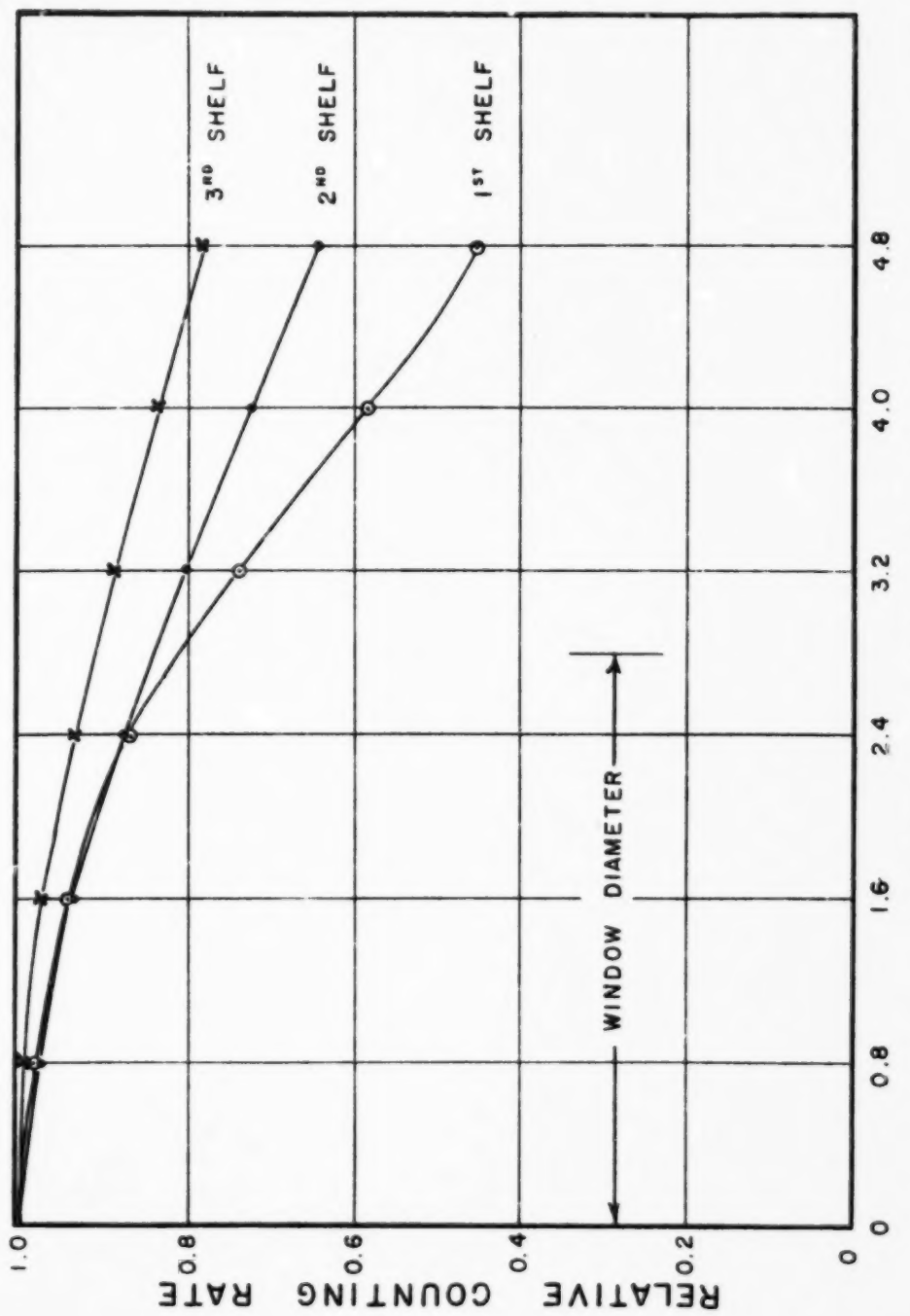

RELATIVE COUNTING RATE
1.0
0.8
0.6
0.4
0.2
0
3RD SHELF
2ND SHELF
1ST SHELF
WINDOW RADIUS
0
4
8
12
16
20
24
HORIZONTAL DISTANCE FROM TUBE AXIS (mm)

FIGURE 3

The counting rate of an extended source relative to the counting rate of an equivalent point source.


3RD SHELF
2ND SHELF
1ST SHELF
WINDOW DIAMETER
RELATIVE COUNTING RATE
1.0
0.8
0.6
0.4
0.2
0
0
0.8
1.6
2.4
3.2
4.0
4.8
SAMPLE DIAMETER (cm)

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